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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 6 :
A61K 7/32, 7/48, C11D 3/00, 3/33, D06M 16/00, 13/342

A1

(11) International Publication Number: WO 97/44006

(43) International Publication Date: 27 November 1997 (27.11.97)

(21) International Application Number: PCT/EP97/02380

(22) International Filing Date: 9 May 1997 (09.05.97)

(30) Priority Data:
196 20 644.8 22 May 1996 (22.05.96) DE

(71) Applicant *(for all designated States except US)*: CIBA SPECIALTY CHEMICALS HOLDING INC. [CH/CH]; Klybeckstrasse 141, CH-4057 Basel (CH).

(72) Inventors; and

(75) Inventors/Applicants *(for US only)*: BACHMANN, Frank [DE/DE]; Lehener Strasse 148, D-79106 Freiburg (DE). OCHS, Dietmar [DE/DE]; Webergasse 11g, D-79650 Schopfheim (DE). UTZ, Ronald [DE/DE]; Riesberg 2, D-79618 Rheinfelden (DE). EHLIS, Thomas [DE/DE]; Ferdinand-Weiss-Strasse 30, D-79100 Freiburg (DE).

(74) Common Representative: CIBA SPECIALTY CHEMICALS HOLDING INC.; Patentabteilung, Klybeckstrasse 141, D-4057 Basel (CH).

(81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, HU, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ARIPO patent (GH, KE, LS, MW, SD, SZ, UG), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).

Published

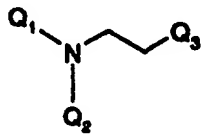
With international search report.

Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.

(54) Title: USE OF NITROGEN-CONTAINING COMPLEXING AGENTS FOR DEODORIZATION AND ANTIMICROBIAL TREATMENT OF THE SKIN AND TEXTILE FIBRE MATERIALS

(57) Abstract

The present invention relates to the use of nitrogen-containing complexing agents for deodorization and antimicrobial treatment of the skin and of textile fibre materials. The complexing agents employed according to the invention have the formula (1), in which Q_1 is $Carb_1$; $Carb_2$; or a radical of the formula (a); Q_2 is hydrogen or $Carb_2$; and Q_3 is $Carb_3$; an amino acid radical; or a radical of the formula (1a), where $Carb_1$, $Carb_2$ and $Carb_3$ independently of one another are the radical of a C_1-C_8-mono- or -dicarboxylic acid and m_1 is 1 to 5. The complexing agents according to the invention show a pronounced bacteriostatic action against Corynebacterium xerosis (bacteria which cause body odour) and are therefore suitable as the antimicrobial active substance in body care compositions and antimicrobial fabric finishing of textile materials.



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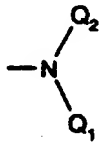

(1a)

$$-(CH_2)_{m_1}-OH \qquad (a)$$

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Use of nitrogen-containing complexing agents for deodorization and antimicrobial treatment of the skin and textile fibre materials

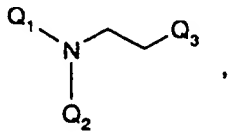
The present invention relates to the use of nitrogen-containing complexing agents for deodorization and antimicrobial treatment of the skin and of textile fibre materials.

It is known that various nitrogen-containing complexing agents, for example ethylenediaminetetraacetic acid (EDTA), nitrilotriacetic acid (NTA), β-alaninediacetic acid (EDETA) or ethylenediaminedisuccinic acid (EDDS) are widely employed in domestic detergents because of their complexing properties.

Surprisingly, it has been found that certain nitrogen-containing complexing agents also have an antimicrobial action against Gram-positive bacteria and are therefore particularly suitable for deodorization and antimicrobial treatment of the human skin and of textile fibre materials.

The present invention therefore relates to the use of nitrogen-containing complexing agents for antimicrobial treatment of the skin and of textile fibre materials.

Compounds which are preferably used according to the invention as complexing agents are those of the formula

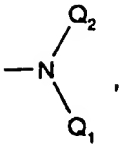
(1) 

,

in which

Q_1, is $Carb_1$; $Carb_2$; or a radical of the formula $-(CH_2)_{m_1}-OH$;

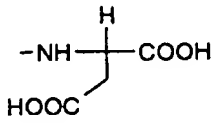
Q_2 is hydrogen or $Carb_2$; and

Q_3 is $Carb_3$; an amino acid radical; or a radical of the formula (1a)  ,

where $Carb_1$, $Carb_2$ and $Carb_3$ independently of one another are the radical of a C_1-C_8-mono- or dicarboxylic acid; and

m_1 is 1 to 5.

Compounds which are particularly preferred here are those of the formula (1) in which the amino acid radical Q_3 has the formula

(1b)  and especially compounds of the formula (1) in which

Q_1 is a monocarboxylic acid; or a radical of the formula $-(CH_2)_{m_1}-OH$;

Q_2 is hydrogen or a monocarboxylic acid; and

Q_3 is formula (1b); or a monocarboxylic acid.

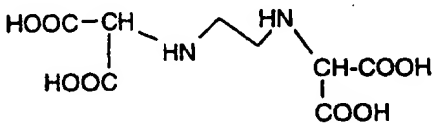
Complexing agents which are of particular interest are those of the formula (1) in which $Carb_2$ and $Carb_3$, independently of one another are the radical of the formula

(1c) $-[(CH_2)]_{n_1}-COOH$,

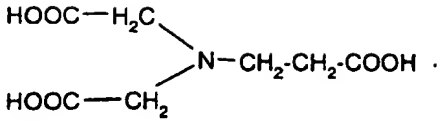
in which

n_1 is 0 to 5.

Complexing agents which are important in practice have the formula

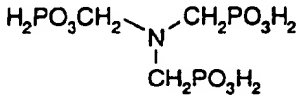
(2) 

or the formula

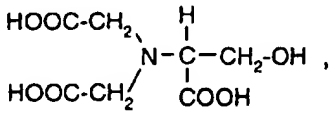
(3) 

Nitrilotriacetic acid (NTA) is furthermore suitable as the complexing agent.

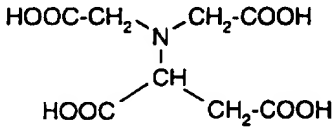
Other examples of complexing agents which can be employed according to the invention are aminotrimethylenephosphoric acid (ATMP) of the formula

(4) 

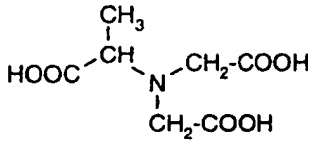
serinediacetic acid (SDA) of the formula

(5) 

asparaginediacetic acid of the formula

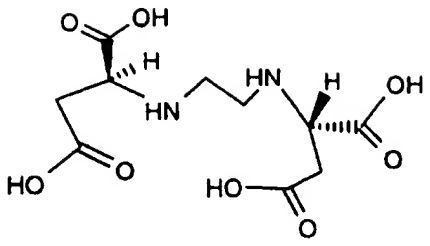
(6)  or

methylglycinediacetic acid (MGDA) of the formula (7)

(7) 

The nitrogen-containing complexing agents employed according to the invention can be employed not only as the acid but also in the form of the water-soluble salts, preferably as lithium, sodium, potassium, ammonium and ethanolammonium salts.

Ethylenediaminedisuccinic acid (EDDS) of the formula (2) has two asymmetric carbon atoms. Various stereoisomeric forms of this compound are therefore possible. The (S,S) configuration of EDDS has the formula

(9) 

An inexpensive chemical synthesis leads to a mixture of the three forms S,S; R,R; and meso-EDDS. However, separation of these stereoisomeric compounds requires a high industrial expenditure. Optically pure (S,S)-EDDS can be prepared with the aid of an Actinomycetes strain (T. Nishikiori et al., Production by Actinomycetes of (S,S)-N,N'-ethylenediaminedisuccinic acid, an inhibitor of phospholipase c; J.Antibiotics 37, 426-427 (1984)).

The purely chemical preparation of the compound of the formula (9) is carried out in a manner known per se, such as is described, for example, by J.A. Neal, N. Rose in Inorganic Chemistry,7, 2405 (1985).

Racemic EDDS can be prepared in accordance with US-A-3 158 635.

The complexing agents according to the invention show a pronounced bacteriostatic action, in particular against Gram-positive bacteria of the skin flora, for example Corynebacterium xerosis (bacteria which causes body odour). They are therefore particularly suitable as the antimicrobial active substance in body care compositions, for example soaps, shampoos, foot care products and, in particular, deodorants, as well as an additive in detergents.

The invention therefore also relates to a body care composition comprising at least one nitrogen-containing complexing agent and carriers or auxiliaries which are tolerated in comsetics.

The body care composition according to the invention comprises 0.01 to 15, preferably 0.5 to 10, % by weight, based on the total weight of the composition, of a nitrogen-containing complexing agent and auxiliaries which are tolerated in cosmetics.

Depending on the form in which the body care composition is present, it also comprises, in addition to the complexing agent, other constituents, for example sequestering agents, dyes, perfume oils, thickeners or consolidating agents (consistency regulators), emmollients, UV absorbers, skin protection agents, antioxidants, additives which improve the mechanical properties, such as dicarboxylic acids and/or Al, Zn, Ca, or Mg salts of C_{14}-C_{22} fatty acids, and, if appropriate, preservatives.

Because of their good water-solubility, the complexing agents according to the invention can be incorporated into the corresponding formulations without problems.

The body care compositions according to the invention can be formulated as a water-in-oil or oil-in-water emulsion, as alcoholic or alcohol containing formulation, as a vesicular dispersion of an ionic or nonionic amphiphilic lipid, as a gel or solid stick or as an aerosol formulation.

As a water-in-oil or oil-in-water emulsion, the auxiliary which is tolerated in cosmetics preferably comprises 5 to 50% of an oily phase, 5 to 20% of an emulsifier and 30 to 90% of water. The oily phase can comprise any oil suitable for cosmetic formulations, for example one or more hydrocarbon oils, a wax, a naturally occurring oil, a silicone oil, a fatty acid ester or a fatty alcohol. Preferred mono- or polyols are ethanol, isopropanol, propylene glycol, hexylene glycol, glycerol and sorbitol.

An anitmicrobial soap has, for example, the following composition:
0.01 to 5% by weight of the compound of the formula (2)
0.3 to 1% by weight of titanium dioxide
1 to 10% by weight of stearic acid
to 100% of soap base, for example the sodium slats of tallow fatty and coconut fatty acid or glycerols.

A shampoo has, for example, the following composition:
0.01 to 5% by weight of the compound of the formula (2),
12.0% by weight of sodium laureth-2-sulfate,
4.0% by weight of cocamidopropylbetaine,
3.0% by weight of NaCl and
water to 100%.

A deodorant has, for example, the following composition:
0.01 to 5% by weight of the compound of the formula (2),
60% by weight of ethanol,
0.3% by weight of perfume oil and
water to 100 %.

The complexing agents according to the invention are furthermore suitable for the treatment of textile fibre materials. The fibre materials are non-dyed and dyed or printed fibre materials, for example of silk, leather, wool, polyamide or polyurethanes, and in particular all types of cellulosic fibre materials. Such fibre materials are, for example, naturally occurring cellulosic fibres, such as cotton, linen, jute and hemp, and cellulose and regenerated cellulose. Textile fibre materials which are preferably suitable are those of cotton.

The following examples serve to illustrate the invention.

Example 1: Determination of the antimicrobial activities of S,S-EDDS, R,R-EDDS, racemate of EDDS and EDETA, EDTA and NTA

Test method: An agar diffusion test is carried out with the following modifications:
Medium: casein-soya flour peptone agar (caso-agar)
Test organisms: Corynebacterium xerosis ATCC 373
Corynebacterium xerosis ATCC 7711
Corynebacterium minutissimum ATCC 23358
Procedure: 500 ml of caso-agar are innoculated with 3.5 ml of an overnight culture of the bacteria, diluted 1:100, and caso plates (18ml) are covered with a layer of about 5 ml of the bacteria-containing agar. After the plates have cooled, holes of diameter 1 cm are stamped out with a cork borer. Each stamped-out hole is filled with in each case 100µl of a

test substance dilution and the plates are incubated at 37°C for 2 days. Double-distilled water is employed as the solvent for all the substances. In the case of EDETA GS, the pH is adjusted to 3.3 by addition of 1 N NaOH. Chemically prepared S,S-EDDS is adjusted to the pH of 5.6 by addition of 1 N NaOH.

Controls: Double-distilled water

The test results are listed in Table 1:

Table 1:		Inhibitory aureola diameter	
Substance	Concen-tration [ppm]	Cory.xerosis ATTC 7711	Cory. xerosis ATTC 373
EDETA	10000	5/5[1]	1/1[1]
S,S-EDDS (prepared chemically)	10000	15/15[1]	10/10[1]
S,S-EDDS (prepared by fermentation)	10000	15/15[1]	10/10[1]
EDTA	10000	2/2	5/5
R,S-EDDS	10000	n.d.	12/13
R,R-EDDS	10000	n.d.	15/15

[1] Slight growth on inhibitory aureolas

The test results show that both EDETA, EDTA and the EDDS prepared by fermentation and chemically (=R,R; S,S; R,S) show a pronounced bacteriostatic action against Coryne-bacterium xerosis.

Examples of formulations having a bacteriostatic action

Example 2: Preparation of a washing powder:

Laurylammonium sulfate	8.0%
Nonionic surfactants	2.9%
Soaps	3.5%
Sodium tripolyphosphate	43.8%
Sodium silicate	7.5%
Magnesium silicate	1.9%
Carboxymethylcellulose	1.2%
EDTA	0.2%
Sodium sulfate	21.2%
EDDS	1%
Water	to 100%

The formulation is prepared as follows:

The solid components are mixed and homogenized in a mortar and stirred with deionized water until a uniform pourable and pumpable paste (slurry) is obtained, which is finally spray-dried.

Example 3: Preparation of a cleansing tonic

Ethanol	20%
Glycerol	5%
PEG-40 hydrogenated castor oil (hydrogenated ethoxylated castor oil)	1%
EDDS	0.5%
Perfume	ad libidum
Water	to 100%

The formulation is prepared as follows:

EDDS is dissolved in ethanol. Under stirring at room temperature PEG-40, glycerol and perfume are added. Finally, the water is added.

Example 3: Preparation of a deodorant stick

Ethanol	20%
Glycerol	30%
Propylene glycol	20%
Ceteareth-25	3%
(= ethoxylated cetyl/stearyl alcohol)	
Sodium stearate	7%
EDDS	0,5%
Perfume	ad libidum
Water	to 100%

The formulation is prepared as follows:
Sodium stearate is melted at 60°C. Propylene glycol, Cetearath-25 and glycerol are added to the melting until a homogeneous clear suspension is obtained. Finally, the suspension is stirred with a EDDS-solution in an alcohol/water mixture at 50°C and cooled slowly.

Example 4: Preparation of soluble EDDS salts and deodorant formulations
S,S-EDDS is obtained by means of microbiological (WO 96/36725) or chemical synthesis (J.A. Neal et al., Inorg.Chem. 7, 2405 (1968)). Racemic EDDS is prepared from maleic anhydride and ethylenediamine (US-A-3 158 635).

A 1 % suspension of racemic EDDS or S,S-EDDS is prepared in water/ethanol (about 7:3) with vigorous stirring. An aqueous solution of NaOH is metered in with an autoamtic titration device until the pH of 7 remains constant for 30 minutes. Any slight milky clouding which occurs is removed by filtering through paper.

By addition of a thickener like hydroxy ethyl cellulose a clear deodorant formulation which is stable at room temperature, comprises about 1% of active substance (based on the tetra-acid) and has a skin-friendly pH is obtained.

If NaOH is replaced by KOH, ammonia or ethanolamine, the corresponding potassium, ammonium and ethanolammonium salts are obtained. Lithium hydroxide, sodium carbonate, sodium bicarbonate or laurylamine can also be employed as the base.

Example 5: Detection of the substantial antimicrobial activity of R,S-EDDS salts on the skin

Formulations (Solutions in 30% ethanol):	1% of R,S-EDDS/sodium salt 1% of R,S-EDDS/amine salt (for the preparation, cf. Example 4)
Medium:	Casein-soya flour peptone agar (caso-agar)
Test organism:	Corynebacterium xerosis ATCC 373

Test method:

Before application of the test solutions, the underarms are washed with a non-antimicrobial soap twice for 1 minute each time. A total of 6ml of test product is then applied to the washed, dry skin of the underarm. Immediately and 2 hours after application of the test products, the EDDS on the skin is extracted by means of discs of filter paper (2cm diameter) moistened in 0.9% NaCl solution (pH: 8.2). For this, the moist filter disc is placed on the treated skin without airbubbles for 4 minutes. The filter discs are subsequently dried at room temperature and then placed on solid agar media with test bacteria.

To prepare the solid agar media, 500 ml of liquid agar are innoculated with 3.5 ml of a 12-16-hour culture, diluted 1:100, of the test bacteria at 47°C and caso plates (18ml) are covered with a layer of about 5ml of the bacteria-containing agar.

After the filter discs have been placed on top, the agar media are incubated for 2 days at 37°C and the inhibition under the filter disc or the inhibitory aureolas of the filter discs is/are then determined.

The test results are listed in Table 2:

Table 2		
Substance		Inhibitory aureola diameter (mm)/ inhibition under the filter disc*
		Coryneb. xerosis ATCC 373
Placebo		0 / 0
R,S-EDDS (sodium salt)		
	immed-iately	5 / 4
	2 hours after application	3 / 4
R,S-EDDS (Amine salt)		
	immed-iately	2,5 / 4
	2 hours after application	2 / 4

* Inhibition under the filter disc:

Explanation:

0 = good growth (no inhibition)

2 = inhibited but clear growth (weak inhibition)

4 = no growth (potent inhibition)

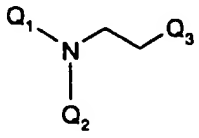
The test results show that a pronounced inhibition of Corynebacterium xerosis is achieved with both test substances.

The test shows that sufficiently high concentrations of EDDS to achieve inhibition of Corynebacterium xerosis are also still present on the skin 2 hours after the last application.

What is claimed is:

1. The use of a nitrogen-containing complexing agent for antimicrobial treatment of the skin and of textile fibre materials.

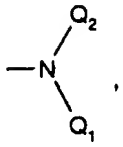
2. The use according to claim 1, wherein a compound of the formula

(1) 

in which

Q_1, is $Carb_1$; $Carb_2$; or a radical of the formula $-(CH_2)_{m_1}-OH$;

Q_2 is hydrogen or $Carb_2$; and

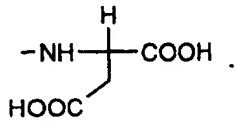
Q_3 is $Carb_3$; an amino acid radical; or a radical of the formula (1a) 

where $Carb_1$, $Carb_2$ and $Carb_3$ independently of one another are the radical of a C_1-C_8-mono- or dicarboxylic acid; and

m_1 is 1 to 5,

is used as the complexing agent.

3. The use as claimed in claim 1 or 2, wherein the amino acid radical Q_3 has the formula

(1b) 

4. The use according to claim 3, wherein

Q_1 is a monocarboxylic acid; or a radical of the formula $-(CH_2)_{m_1}-OH$;

Q_2 is hydrogen or a monocarboxylic acid; and

Q_3 is formula (ib); or a monocarboxylic acid.

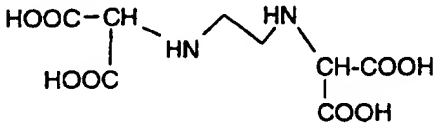
5. The use according to claim 1 or 2, wherein $Carb_2$ and $Carb_3$, independently of one another are the radical of the formula

(1c) $-[(CH_2)]_{n_1}-COOH$,

in which

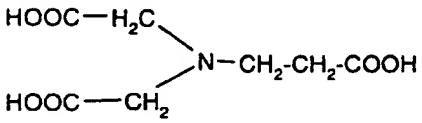
n_1 is 0 to 5.

6. The use according to any one of claims 1 to 4, wherein the compound of the formula

(2) 

is employed as the complexing agent.

7. The use according to any one of claims 1 to 4, wherein the compound of the formula

(3) 

is employed as the complexing agent.

8. The use according to claim 1, wherein nitrilotriacetic acid is used as the complexing agent.

9. The use of a complexing agent according to any one of claims 1 to 8, wherein the complexing agent is also in the form of one of its water-soluble salts.

10. The use according to claim 9, wherein the complexing agent is present in the form of its lithium, sodium, potassium, ammonium or ethanolammonium salt.

11. The use of a complexing agent according to any one of claims 1 to 10 as an antimicrobial active substance against Gram-positive bacteria.

12. The use of a complexing agent according to any one of claims 1 to 11 in body care compositions.

13. A body care composition comprising a nitrogen-containing complexing agent according to claim 1.

14. A body care composition according to claim 13 in the form of a soap, a shampoo or a deodorant.

15. The use of a complexing agent according to any one of claims 1 to 10 in textile fibre materials.

INTERNATIONAL SEARCH REPORT

Inter ional Application No
PCT/EP 97/02380

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 A61K7/32 A61K7/48 C11D3/00 C11D3/33 D06M16/00
D06M13/342

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 A61K C11D D06M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	FR 2 202 698 A (BEECHAM GROUP) 10 May 1974 see claims 1-3,9 see page 3, line 15-28 see page 4, line 19-21 see page 4, line 27-35 see page 6, line 32-37 see examples 3,4 ---	1-6,9-15
X	EP 0 312 700 A (M.CURTI) 26 April 1989 see claims 1,5,7 ---	1,2,5,9, 10,12-14
X	EP 0 328 091 A (H.-G. KAISER) 16 August 1989 see claims 1,2,11,12 see column 1, line 18-24 see column 2, line 9-13 see column 3, line 16-32 --- -/--	1-4,7-13

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

* Special categories of cited documents :

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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"&" document member of the same patent family

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Date of the actual completion of the international search	Date of mailing of the international search report
16 September 1997	26.09.97
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+ 31-70) 340-2040, Tx. 31 651 epo nl, Fax (+ 31-70) 340-3016	Authorized officer Peeters, J

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INTERNATIONAL SEARCH REPORT

Inter ional Application No
PCT/EP 97/02380

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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Inte: ɔnal Application No
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